Supporting Information

Engineering redox-active benzo[1,2-b:4,5-b']dithiophene-based conjugated polymers: tuning porosity and linker architecture for high-performance supercapacitors

Yousra. M. Nabil,^{1,2} Shimaa Abdelnaser,^{1,2} Ahmed A. K. Mohammed,² Shiao-Wei Kuo,¹ and Ahmed F.

M. EL-Mahdy*1,2

[1] Department of Materials and Optoelectronic Science, National Sun Yat-Sen University,

Kaohsiung 80424, Taiwan.

[2] Chemistry Department, Faculty of Science, Assiut University, Assiut 71516, Egypt.

Corresponding authors: Ahmed F. M. EL-Mahdy, +886-986-404-402, E-mail: <u>ahmedelmahdy@mail.nsysu.edu.tw</u>

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S1. Materials.

Tetrakis(triphenylphosphine)palladium (Pd(PPh₃)₄) (99 %), N-bromosuccinimide (NBS) (99 %) were obtained from Acros, while 1,4-Phenylenediboronic acid (99% purity) was obtained from TCI America. 3.5-triethynylbenzene (99% purity) and 1.4-diethynylbenzene were obtained from Leyan.com, while benzene-1,3.5-triboronic acid (99% purity) was obtained from Bidefarm.com. potassium carbonate (K2CO3) was delivered from Combi-Blocks. Triethyl amine (TEA), Cupper iodide, and N,N-Dimethylformamide (DMF) were provided from Sigma Aldrich.

S2. Characterization.

Fourier-transform infrared spectroscopy (FTIR) was conducted with using a Bruker Tensor 27 FTIR spectrophotometer and the standard KBr plate technique; 32 scans were obtained at a resolution of 4 cm⁻¹. Solid state nuclear magnetic resonance (SSNMR) spectroscopy was performed with a Bruker Avance 400 NMR spectrometer along with a Bruker magic-angle-spinning (MAS) probe, conducting 32,000 scans. Thermogravimetric analysis (TGA) was conducted with a TA Q-50 analyzer while nitrogen was flowing. The samples were enclosed in a Pt cell and heated from 40 to 800 °C at a rate of 20 °C min-1 under a nitrogen atmosphere with a flow rate of 50 mL min⁻¹. Nitrogen adsorption-desorption experiments were performed utilizing a BelSorp max device. Prior to assessing gas adsorption, the prepared samples (50 mg) underwent a washing process with anhydrous tetrahydrofuran for 24 hours through Soxhlet extraction. The solvent underwent filtration, and the samples were subjected to activation for 10 hours under pressure at a temperature of 150 °C. Subsequently, the samples were utilized for gas adsorptiondesorption measurements at 77 K within the range of 0 to 1 atm. The Brunauer-Emmett-Teller (BET) method was used to calculate their specific surface areas. The pore distributions were derived from the sorption data by employing quenched solid state density functional theory scanning electron microscopy (SEM) was performed using a JEOL JSM-7610F scanning electron microscope. Samples were subjected

to Pt sputtering for 100 seconds before observation. Transmission electron microscopy (TEM) was performed using a JEOL-2100 scanning electron microscope, operating at 200 kV.

S3. Synthetic Procedures.

S3.1. Synthesis of 2,6-dibromobenzo[1,2-b:4,5-b']dithiophene-4,8-dione (DTDO-2Br).

Using a 250 mL two-neck flask, 2,5-dibromo-N,N-dimethylthiophene-3-carboxamide (2.00 g, 6.4 mmol) was dissolved in 50 mL of dry tetrahydrofuran (THF) and cooled to -78 °C while maintained under a nitrogen environment. A solution of n-butyllithium (n-BuLi, 2.50 mL, 6.4 mmol, 2.5 M in hexane) was added to the previously prepared cold suspension. The reaction mixture was stirred at -70 °C for 4 hours and then allowed to attain room temperature. Saturated aqueous solution of ammonium chloride (20 mL) was added to terminate the reaction. The resulted precipitate was collected filtration. The crude product underwent purification through silica gel column chromatography using chloroform as the eluent, yielding DTDO-2Br as an orange solid (1.06 g, 44% yield).



Scheme S1. Synthesis of ,6-dibromobenzo[1,2-b:4,5-b'] dithiophene-4,8-dione (DTDO-2Br).

S3.2. General preparation of Ph-DTDO porous

A Mixture of DTDO-2Br (0.4 mmol), Ph-3B(OH)₂ (0.26 mmol), potassium carbonate (547 mg), and Pd(PPh₃)₄ (0.038 mmol) was added into a 50 mL Pyrex reaction tube and vacuumed for 10 minutes. Subsequently, 10 mL of DMF and 2 mL of water were included. The tube underwent three cycles of

freeze–pump–thaw, then was purged with nitrogen to eliminate any residual oxygen. The tube was maintained at 130 °C for three successive days and then allowed to cool to 25 °C. finally, the reaction mixture was subsequently poured into ice water, and the resultant solid was filtered, washed thrice with methanol and tetrahydrofuran and Acetone then dried at 60°C overnight under vacuum to yield Ph-DTDO porous polymer as a black solid.



Scheme S2. Synthesis of Ph-DTDO porous conjugated polymer.

S3.3. General preparation of Ph-DTDO linear conjugated polymers

A Mixture of DTDO-2Br (0.4 mmol), Ph-2B(OH)₂ (0.4 mmol), potassium carbonate (547 mg), and Pd(PPh₃)₄ (0.038 mmol) was added into a 50 mL Pyrex reaction tube and vacuumed for 10 minutes. Subsequently, 10 mL of DMF and 2 mL of water were included. The tube underwent three cycles of freeze–pump–thaw, then was purged with nitrogen to eliminate any residual oxygen. The tube was maintained at 130 °C for three successive days and then allowed to cool to 25 °C. finally, the reaction mixture was subsequently poured into ice water, and the resultant solid was filtered, washed thrice with methanol and Acetone then dried at 60°C overnight under vacuum to yield Ph-DTDO linear conjugated polymer as a black solid.



Scheme S3. Synthesis of Ph-DTDO linear conjugated polymer.

S3.4. General preparation of TEPh-DTDO porous conjugated polymers

A mixture of DTDO-2Br (0.4 mmol), TEPh $3C \equiv C$ (0.26 mmol), cupper iodide (4 mg), and Pd(PPh₃)₄ (0.02 mmol) was placed under vacuum for 10 minutes in a 50 mL Pyrex reaction tube. 5 mL of TEA and 5 mL of DMF were then added. The tube was evacuated with nitrogen after three cycles of freeze-pump-thaw to get rid of any last remnants of oxygen. The tube was heated at 130 °C for three days, then cooled to 25 °C. Subsequently, the reaction mixture was poured into ice water, and the resulting solid was filtered and washed three times with methanol, tetrahydrofuran, and acetone, then dried overnight under vacuum at 60°C to produce TEPh-DTDO porous conjugated polymer as a brown solid.



Scheme S4. Synthesis of TEPh-DTDO porous conjugated polymer.

S3.5. General preparation of DEPh-DTDO linear conjugated polymers

A mixture of DTDO-2Br (0.4 mmol), DEPh 2C=C (0.4 mmol), cupper iodide (4 mg), and Pd(PPh₃)₄ (0.02 mmol) was placed under vacuum for 10 minutes in a 50 mL Pyrex reaction tube. 5 mL of TEA and 5 mL of DMF were then added. and the tube was purged with nitrogen following three freeze-pump-thaw cycles to eliminate any remaining oxygen. The tube was heated to 130 °C for a duration of three days and then allowed to cool to 25 °C. After that, the reaction mixture was transferred into ice water, and the formed solid was filtered and washed three times using methanol and acetone, followed by drying overnight under vacuum at 60°C to yield DEPh-DTDO linear conjugated polymer as a brown solid.



Scheme S5. Synthesis of DEPh-DTDO linear conjugated polymer.

S4. Spectral Analyses of DTDO-2Br Monomer.



Figure S1. FTIR spectrum of DTDO-2Br.



Figure S2. ¹H-NMR spectrum of DTDO-2Br.



Figure S3. ¹³C-NMR spectrum of DTDO-2Br.

S5. conjugated polymers suspension in DMF solvent.



Figure S4. illustrates that both crosslinked and linear DTDO-conjugated polymer s are insoluble in DMF.

S6. Spectral Analyses of conjugated polymers.



Figure S5. FTIR spectrum of Ph-3B(OH)₂, DTDO-2Br, Ph-DTDO porous conjugated polymer.



Figure S6. FTIR spectrum of Ph-2B(OH)₂, DTDO-2Br, Ph-DTDO linear conjugated polymer.



Figure S7. FTIR spectrum of TEPH 3C=C, DTDO-2Br, TEPh-DTDO porous conjugated polymer.



Figure S8. FTIR spectrum of DEPh-2C≡C, DTDO-2Br, DEPh-DTDO linear conjugated polymer.

S7. XPS analysis of conjugated polymers



Figure S9. (a) Chemical structures of DTDO-based conjugated polymers and (b) XPS spectra of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear conjugated polymers.

S8. XRD analysis of conjugated polymers



Figure S10. (a) Powder XRD Patterns of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear polymers. (b-e) Geometrical structures and interlayer distances of (b) Ph-DTDO porous, (c) Ph-DTDO linear, (d) TEPh-DTDO porous, and (e) DEPh-DTDO polymers.



S9. TEM, SEM, and EDS elemental mapping of conjugated polymers

Figure S11. (a) TEM and SEM image of Ph-DTDO porous, (b) TEM and SEM image of Ph-DTDO linear, (c) TEM and SEM image of TEPh-DTDO porous, and (d) TEM and SEM image of DEPh-DTDO linear recorded at different scales.



Figure S12. (a-d) EDS elemental mapping images for Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear respectively.

S10. DFT calculations.

We performed density functional theory (DFT) calculations on the linear and porous polymers to reveal their structure and gain insights into their properties. The ground state geometry of all structures was optimized using DFT with the B3LYP functional with the D3BJ correction and the basis set 6-31G(d). This dispersion correction is necessary to account for long-range and noncovalent interactions. Several conformers were considered for the ground state geometry, and the one with lowest energy was selected. The lack of imaginary frequencies proved that these geometries are true minima. All geometry optimizations were performed using the Gaussian 16 software. Molecular electrostatic potential maps were generated using the Multiwfn software.



Figure S13. (a-d) The HOMO distribution, LUMO distribution, and the band gab energy values Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear.

S11. Electrochemical Analysis in Three-Electrode System.

Working electrode cleaning:

Prior to use, the glassy carbon electrode (GCE) was polished multiple times with 0.05-µm alumina powder

before use. Each polishing step followed by a rinse with EtOH, a 5-minute sonication in a water bath,

another EtOH wash, and then air drying.

Electrochemical Assessment.

Electrochemical experiments were performed in a three-electrode configuration using a Zahner potentiostat with a (3 M) KOH aqueous electrolyte. The GCE, featuring a diameter of (5.61 mm) and an area of (0.2475 cm²), functioned as the working electrode. A platinum wire served as the counter electrode, and a Hg/HgO electrode (RE-61AP, BAS) functioned as the reference electrode; all potentials are expressed relative to Hg/HgO. The GCE was modified using conjugated polymer slurries formulated with minor changes to previously established methods.¹⁻⁴ The slurries were prepared by dispersing 70 wt% conjugated polymer, 20 wt% multi-walled carbon nanotubes (MW-CNT), and 10 wt% Nafion in a solution of 0.2 mL ethanol and 0.8 mL deionized water, then subjected to sonication for one hour. A 5 µL portion of this slurry was deposited onto the surface of the GCE and permitted to air-dry for 30 minutes before using. The electrochemical performance was assessed via cyclic voltammetry (CV) at scan rates between 5 and 200 mV s⁻¹, and galvanostatic charge-discharge (GCD) measurements within a potential range of -0.7 to 0 V for all of Ph-DTDO porous, TEPh-DTDO porous, DEPh-DTDO linear, and -0.7 to -0.2 for Ph-DTDO linear, versus Hg/HgO at varying current densities from 0.5 to 20 A g⁻¹ in a 3 M KOH aqueous electrolyte solution. The specific capacitance was calculated from galvanostatic chargedischarge experiments employing the following equation:^{4, 5}

$Cs = (I\Delta t)/(m\Delta V)$ (S1)

Cs (F/g) denotes the specific capacitance of the supercapacitor, I (A) represent the discharge current, ΔV refers to the potential window in (V), Δt (s) indicates the discharge duration, and m (g) stands for the mass of CONJUGATED POLYMER loaded on the electrode. The energy density (E, Wh/kg) and power density (P, W/kg) determined using equations (S2 and S3):^{5, 6}

$$E = 1000C(\Delta V)^2 / (2*3600)$$
(S2)

P = E/(t/3600) (S3)



position at -0.48 and -0.51 V.

Figure S14. (a) CV, and (b) GCD curves for Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear at scan rate 5 mV s⁻¹.



Figure S15. (a) CV curves, (b) GCD curves, and (c) specific capacitance of MW-CNT.



Figure S16. The suggested redox reactions of DTDO redox active unit.



Figure S17. CV profiles of (a) Ph-3B(OH)₂, (b) Ph-2B(OH)₂, (c) DTDO-2Br, (e) TEPh 3C=C, and (e) DEPh 2C=C.



Figure S18. The equivalent circuit of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear conjugated polymers.

S12. Electrochemical Analysis in Two-Electrode Symmetric Supercapacitor System.

The slurry was made up of conjugated polymer (70 wt. %), carbon black (20 wt. %), and Nafion (10 wt. %), was applied to a Kuraray carbon paper (0.1 ± 0.01 mm thick) with a functional area of 1 cm × 1 cm and subsequently dried at 100 °C overnight in a vacuum oven. The bulk loading of active material on the current collector was 1.00 mg cm⁻². A filter paper was employed to separate the two operating electrodes

and saturated with a 3 M of an aqueous potassium hydroxide solution. The specific capacitance was calculated using galvanostatic charge discharge experiments with the following equation S4:^{2, 7}

$$Cs = 2 x (I\Delta t)/(m\Delta V)$$
 (S4)

(F/g) signifies the specific capacitance, I (A) represent the discharge current, ΔV (V) indicates the potential window, and m (g) refers to the mass of conjugated polymer on the electrode. A factor 2 is multiplied due to the formation of series capacitance in a symmetrical supercapacitor system.

The coulombic efficiency (%) was determined using the following equation.⁸

$$\eta (\%) = (\Delta t d / \Delta t c) \times 100$$
 (S5)

 Δ td, Δ tc are the discharging time and charging time, respectively.



Figure S19. Nyquist plot of Ph-DTDO porous conjugated polymer-based SC device.

S13. Tables.

Table S1. XPS fitting positions of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO

Samples			C species				S species		
		С-С	C=C	C≡C	C—S	С=0	S2s	S2p _{3/2}	S2p _{1/2}
conjugated polymers	Ph-DTDO porous	284.62	283.79		284.05	286.56	227.84	163.58	164.77
	Ph-DTDO linear	284.50	283.49		284.02	286.50	227.45	163.29	164.49
	TEPh-DTDO porous	284.05	283.74	284.98	284.04	286.66	227.64	163.47	164.71
	DEPh-DTDO linear	284.43	283.63	284.47	284.10	286.54	227.54	163.33	164.57

linear conjugated polymers.

Table S2. XPS fitting ratio of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear conjugated polymers.

Samples			C species				S species		
		С-С	C=C	C≡C	C—S	С=0	S2s	S2p _{3/2}	S2p _{1/2}
conjugated polymers	Ph-DTDO porous	11.93%	37.40%		32.62%	18.02%	100%	65.70%	34.29%
	Ph-DTDO linear	10.72%	39.24%		34.86%	15.16%	100%	63.87%	36.12%
	TEPh-DTDO porous	15.28%	34.55%	11.66%	24.69%	13.80%	100%	64.04%	35.95%
	DEPh-DTDO linear	11.23%	43.90%	5.55%	24.93%	14.36%	100%	67.49%	32.50%

Table S3. BET parameters of the synthesized conjugated polymers.

Conjugated polymer	S _{BET} (m ² g ⁻¹)	Pore volume (cm ³ g ⁻¹)	Pore size (nm)
Ph-DTDO porous	62.7	0.36	1.65–3.41
TEPh-DTDO porous	92.3	0.43	1.58-3.77

Table S4. TGA Td5, Td10, and Char yield of DTDO- conjugated polymers.

Conjugated polymer	T _{d5} (°C)	T _{d10} (°C)	Char yield (%)
Ph-DTDO porous	425.50	537.94	63.50
Ph-DTDO linear	536.64	583.71	67.93
TEPh-DTDO porous	359.64	474.66	57.76
DEPh-DTDO linear	310.20	434.57	57.14

No	Polymers	S _{BET} (m ² g ⁻¹)	Capacitance (F g ⁻¹)	Stability	Ref.
1	BTPP-DBTh	388	143.27, at 0.5 A g ⁻¹	87.86 % after 10, 000 cycles, at 0.5 A g ⁻¹	2
2	COF/rGO	246	269, at 0.5 A g ⁻¹	96% after 5,000 cycles, at 8 A g^{-1}	9
3	BF-DTDO	410	95.62, at 0.5 Ag ⁻¹	$\begin{array}{l} 69.69\% after 10000 \\ cycles, at 10 \ A \ g^{-1} \end{array}$	10
4	BF-Ph-DTDO	336	288.8 at 0.5 Ag-1	76.58% after 10000 cycles, at 10 A g-1	10
5	TAT-CMP-1	106	141, at 1 A g ⁻¹	$\begin{array}{ccc} 95\% & after & 10000 \\ cycles, at 10 \ A \ g^{-1} \end{array}$	11
6	TAT-CMP-2	141	183, at 1 A g ⁻¹	$\begin{array}{c} 83\% after 10000 \\ cycles, at 10 \ A \ g^{-1} \end{array}$	11
7	TPA -Py	1470	78, at 1 A g ⁻¹	96.7% after 2000 cycles, at 10 A g^{-1}	12
8	TPA-Bz	773	55.1, at 1 A g ⁻¹	95.2% after 2000 cycles, at 10 A g^{-1}	12
9	BT-PDI	161.46	196, at 1 A g ⁻¹	76% after 5000 cycles, at 5A g^{-1}	13
10	c-DDSQ-MDA- BMI	826	73.6, at 0.5 A g^{-1}	$\begin{array}{c} 86.4\% after 5000 \\ cycles, at 10 \ A \ g^{-1} \end{array}$	14
11	cNPIMEA- TB_80	1966	46, at 1 A g ⁻¹	$\begin{array}{ccc} 90\% & after & 10000 \\ cycles, at 1.7 \ A \ g^{-1} \end{array}$	15
12	CoPc- CMP/CNTs-2	150.69	107.2, at 1 A g ⁻¹	89.2% after 1350, at 10 A g ⁻¹	16
13	GH-CMP	219.2	182.7, at 0.5 A g ⁻¹	92.6 % after 10 000 cycles, at 2 A g^{-1}	17
14	CAP-2	594	233, at 1 A g ⁻¹	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	18
15	An-TPP POP	9	38.12, at 1 A g ⁻¹	$\begin{array}{c} \mbox{78.52 \ \%after \ 10,000} \\ \mbox{cycles, at 10 A g}^{-1} \end{array}$	19
16	An-TPA POP	18.52	117.7, at 1 A g ⁻¹	81.55 %after 10,000 cycles, at 10 A g^{-1}	19
17	Ph-DTDO porous	62.7	842.40, at 0.5 A g ⁻¹	98.78%, after 6000 cycles, at 30 A g^{-1}	This work
18	Ph-DTDO linear		429.2 at 0.5 A g ⁻¹	75.07%, after 6000 cycles, at 30 A g^{-1}	This work
19	TEPh-DTDO porous	92.3	399.6, at 0.5 Ag ⁻¹	92.06%, after 6000 cycles, at 30 A g^{-1}	This work

Table S5. Comparison of the specific surface area/specific capacitance of DTDO-based conjugated polymers with those of conjugated porous polymers that have been previously reported for supercapacitor applications.

20	DEPh-DTDO	 327.1, at 0.5 Ag ⁻¹	89.70%,	after	6000	This work
	linear		cycles, at	30 A g	-1	

Table S6. The fitting values of Nyquist plots of Ph-DTDO porous, Ph-DTDO linear, TEPh-DTDO porous, and DEPh-DTDO linear conjugated polymers.

Polymer	Rs	Rct
Ph-DTDO porous	1.34	79
Ph-DTDO linear	2.98	95
TEPh-DTDO porous	3.25	102
DEPh-DTDO linear	4.47	299

Table S7. A comparison of the electroconductivity Ph-DTDO-based conjugated polymer with other polymeric materials use analogous redox-active materials.

No	Polymers	Electroconductivity (S cm ⁻¹)	Ref.
1	BT-CMP	6.96×10 ⁻⁸	20
2	DT-P	3×10 ⁻³	21
3	CPFs	1.3×10 ⁻²	22
4	Ph-DTDO porous	5.88	This work
5	Ph-DTDO linear	4.95	This work
6	TEPh-DTDO porous	4.57	This work

7	DEPh-DTDO linear	2.51	This work
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Table S8. A comparison of the specific capacitance of the Ph-DTDO porous conjugated polymer supercapacitor device with that of symmetrical supercapacitor devices that have been previously documented and use analogous redox-active materials.

No	Polymers	S _{BET} (m ² g ⁻¹)	Capacitance (F g ⁻¹)	Stability	Ref.
1	PTPA@MWNT-4	484	216, at 1 A g ⁻¹	71% after 6,000 cycles, 2 A g ⁻¹	23
2	COF/rGO	246	269, at 0.5 A g^{-1}	96% after 5,000 cycles, at 8 A g^{-1}	9
3	AC in 1M Na2SO4	1601	94, at 0.2 A g ⁻¹	98.9% after 1000 cycles, at 1 A g^{-1}	24
4	PAQTA	331	129, at 1 A g ⁻¹	98% after 2000 cycle, at 2 A g^{-1}	25
5	BC-PT CMP	478	107, at 1.2 A g ⁻¹	97.69% after 5000 cycles, at 10 A g^{-1}	26
6	SDBPy	1200	176, at 0.25 A g ⁻¹	92.6% after 10000 cycles, at 0.25 A g ⁻¹	27
7	MWCNT@SACMP	514	254, at 1 A g ⁻¹	84.38%, after 6000 cycles at 2 A g^{-1}	28
8	Ph-DTDO porous	62.7	428.21, at 0.43 A g^{-1}	88.51% after 4000 cycles, at 1.73 A g ⁻¹	This work

S14. References.

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